Constructed Wetlands for Treating Processed Mine Water
- an Irish Case Study

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Abstract

Mine wastewater, characteristically elevated in metals and sulfate, is conventionally treated with costly chemical applications. The development of passive treatment systems, employing both biotic and abiotic processes, has been recognized as an economically feasible, ecologically acceptable technology in the last decade. However, to-date most of these passive systems have been applied to abandoned mine waters for the primary purpose of increasing pH and removing metals. Two experimental-scale treatment wetlands were constructed and monitored at an active lead/zinc mine (Tara Mines) in Ireland, to treat alkaline mine seepage with elevated sulfate (and also metal) levels. Each system comprised three 12 m^2 (2 m depth) in-series surface-flow cells *viz.*, inflow, vegetated and outflow and contained spent mushroom substrate (SMS). Typical aqueous concentrations of 830 mg L^{-1} sulfate, 0.15 mg L^{-1} lead and 2.0 mg L^{-1} zinc entered the treatment wetlands at a flow rate of c. 650 mL min^{-1}. Anaerobic substrates, in which sulfate-reducing bacteria were indigenous, were conducive to biological reduction of sulfate to sulfide. Sulfide subsequently precipitated with metal cations. Monitoring of these wetlands over a two-year period showed successful (maximum) removal of sulfate (29 g m^{-2} day^{-1} (69%)), lead (6.6 mg m^{-2} day^{-1} (64%)), and zinc (70 mg m^{-2} day^{-1} (98%)). These contaminants were somewhat associated with plant roots but more significantly so with the substrates. All of the interacting processes within the wetland ecosystems, responsible for decontamination of the wastewater, are currently being elucidated and quantified using a system dynamic model. The communities of colonizing macroinvertebrates, macrophytes, algae and microorganisms also contributed to the development of a diverse ecosystem.

Additional Key Words: sulfate, heavy metals, mine water, vegetation

Introduction

Mine wastewater

Water quality of run-off and leachate derived from metalliferous mine tailings is generally not compliant with Irish (O'Leary, 1996) or international discharge standards (Allan, 1995, Novotny, 1995). It is characteristically elevated in metal and sulfate concentrations resulting from ore processing. Most zinc (sphlaerite), lead (galena), iron (marcasite, pyrite) and cadmium (greenockite) ore bodies contain high proportions of sulfur. Additionally, sulfuric acid used during extraction of these ores contributes to elevated sulfate levels in the discharge water (Tara Mines, 1996).

Treatment of mine wastes

Since 1982, the decontamination of mine wastes employing biological passive treatment technologies has developed substantially in lieu of conventional chemical applications (Nawrot, 1994,

Saunders et al., 1996, Gusek & Wildeman, 2002). Typically, wetlands are incorporated into this treatment process and are referred to as 'constructed wetlands'. The success in using wetlands for mine waste remediation efforts has been demonstrated in many capacities; their self-renewing capabilities, good quality performance and ecological benefits (Debusk et al., 1996, Beckett, 1999, O'Sullivan et al., 1999). Many different types of constructed wetlands are discussed in detail elsewhere (Vymazal et al., 1999, Mitsch & Gosselink, 2000, Gusek & Wildeman, 2002).

By comprehending the complex and dynamic biogeochemical properties of wetlands, the behavior of metal and sulfate contaminants in these ecosystems can be biotically and abiotically

controlled (Dunbabin & Bowmer, 1992, Fortin *et al*., 1995). In moderate climates (such as that prevailing in Ireland), anaerobic wetland substrates are conducive to biological sulfate reduction (Eqn. 1). Populations of sulfate-reducing bacteria were indigenous in the spent mushroom substrate (SMS) used in the treatment wetlands at Tara Mines (O'Sullivan, 2001). This substrate was also rich in organic material, which was simultaneously oxidized during microbial reduction of sulfate. Organic material was renewed cyclically through annual plant decomposition. Metal cations, such as Zn and Pb, bind with reduced sulfides under alkaline conditions to produce metal-sulfide precipitates, carbon-dioxide and water (Eqn. 2).

Study site

Tara Mines Ireland (a subsidiary of Outokumpu-Zinc) is reputedly the largest producer of zinc in Europe. The local geology of the mine near Navan, Co. Meath in Ireland comprises Lower Carboniferous calcite ($CaCO_3$) and dolomite ($CaMgCO_3$). This rock buffers the spent wastewater discharged from the mine, via carbonate dissolution (Eqn. 3), to a pH of approximately 7.8 (O'Leary, 1996). Wastewater is then stored in large tailings impoundment dams 5.6 km north of the mine (53º 42' N, 06º 43' W). Some of this wastewater is fed to the experimental treatment wetlands described here.

Equations
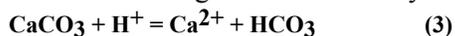
Biological sulfate reduction generates alkalinity

$$2CH_2O + SO_4^{2-} = H_2S + 2HCO_3^- \qquad (1)$$

Metal-sufide formation produces carbon-dioxide

$$M^{2+} + H_2S + 2HCO_3^- = MS + 2H_2O + 2CO_2 \quad (2)$$

where, M^{2+} are metal cations (e.g. Zn, Pb, Fe) and MS are metal-sulfides.

Carbonate dissolution generates alkalinity

$$CaCO_3 + H^+ = Ca^{2+} + HCO_3 \qquad (3)$$

Materials and Methods

Design specifications and Experimental set-up

Two experimental treatment systems were constructed at Tara Mines in 1997, specifically to treat sulfate and metals in wastewater. They are the only treatment wetlands of this kind in Europe at the time of this publication. Most applications employing microbes to remove contaminants from wastewater have focused on metal removal (Dunbabin and Bowmer, 1992, Debusk *et al*., 1996, Song *et al*., 1998, Vymazal *et al*., 1999), while the research at Tara Mines was concerned primarily with treating sulfate from the alkaline mine water. Each system comprises three 16 m² (2m depth) in-series surface flow (SF) ponds, containing an inflow, vegetated cell and outflow, which were filled with spent mushroom substrate and a fine inert grit, by volume of 1:6 (inflow and outflow, 50cm depth) and 1:3 (vegetated, 1m depth). Refer to Figure 1 for details.

Measurements and Analyses

Sulfate was measured by Ion Chromatography using Dionex instrumentation (QIC analyzer, automated sampler and 4400 integrator) equipped with a separator column and conductivity cell. Vegetation and substrate samples were oven-dried at 60 ºC for 5 days, ground using a mortar and pestle and sieved through a 2,000 μm aluminum sieve. Samples were then digested overnight in strong acid under high temperature, using Teflon™ bombs as described by Beining and Otte (1996). Lead, zinc and iron were analyzed in the vegetation and substrate extracts and in water using a Unicam 929 Atomic Absorption Spectrophotometer supporting SOLAAR ATI software. Concentrations were log-transformed to obtain homogeneity of variance. Data were analyzed using SAS (v 6.1) to test for significant differences by conducting a series ANOVAs.

Results

Water characteristics

Water quality discharged from Tara Mines was generally comparable with other lead/zinc alkaline wastewaters (Allan, 1995, Gao and Bradshaw, 1995). However, concentrations of metals supplied to the treatment wetlands were typically lower

than that of other mine waters. This likely results from some metal precipitation within the storage tailings dam and is reported elsewhere (Finnegan, 1998, Song *et al*., 1998). Water quality for net alkaline mine drainage from an abandoned lead/zinc facility at Mayer Ranch in northeastern Oklahoma (USA) is also given in Table 1. Data sets from both Tara Mines and Mayer Ranch are being incorporated into a mass balance budget model currently being developed.

Table 1. Typical water quality data for mine drainage at Tara Mines, Ireland and Mayer Ranch, Oklahoma. Water is net alkaline in both sites and concentrations are given in mg L^{-1}. Values in brackets for Tara Mines represent metal concentrations supplied to the experimental treatment wetlands.

Parameter	Tara Mines	Mayer Ranch
Cond. (μS cm^{-1})	2000	3600
pH	7.5 - 8.1	5.9 - 6.5
Air temperature (^{o}C)	3 – 21	-7 – 33
	Concentration (mg L^{-1})	
SO_4	900	3000 - 5000
Alkalinity ($CaCO_3$)	350	414
Pb	0.2 (0.15)	0.02
Zn	5.0 (2.0)	11
O_2	8.5	0.1

Sulfate mass removal
Removal of sulfate in system 2 ranged between 1-29 g m^{-2} day^{-1}, while generally lower removal rates of 1-18 g m^{-2} day^{-1} (with the exception of 42 g m^{-2} day^{-1} in September 1998) were calculated for system 1 (Table 2). The treatment systems had reached equilibration in October 1998 (determined from preliminary data), approximately 1 year following construction. These data (Nov. 1997 - Oct. 1998) were discussed in detail elsewhere (O'Sullivan *et al*. 1999, O'Sullivan, 2001) but are also included within the context of this paper to highlight differences in operation of the systems before and after equilibration. Typically, more sulfate was removed following equilibration of the treatment systems. Reduction in sulfate concentrations was significant in both systems at the p<0.001 level. Furthermore, differences in concentration as a function of sampling date were

also significant for system 1 (p<0.01) and for system 2 (p<0.001).

Table 2. Sulfate removal rates (g m^{-2} day^{-1}) from water throughout the monitoring period. Values are calculated from differences in concentrations between water entering and exiting the treatment systems (i.e. from the source to the outflow) as a function of the average flow rate (1.5 L min^{-1}), n=5. Sulfate removal was calculated from concentration differences between the inflow and outflow cells in earlier sampling dates (i.e. not throughout the complete water cycle) as indicated by *. The calculation is summarized below (Eqn. 4). Equivalent percentage removals are indicated in parenthesis. A negative value (16 Sep. 1998 for system 2) indicates that there was a net export of sulfate from the system on that sampling date.

SAMPLING	SYSTEM 1	SYSTEM 2
	g m^{-2} day^{-1}	g m^{-2} day^{-1}
01 Feb. 1998	0 {2}*	1 {4}*
31 Aug. 1998	12 {24}	6 {6}*
16 Sep. 1998	42 {81}	-1 {0}*
05 Oct. 1998	0 {0}*	10 {34}*
19 Oct. 1998	5 {14}	4 {11}
16 Nov. 1998	18 {62}	13 {45}
21 Dec. 1998	11 {38}	29 {69}
01 Feb. 1999	9 {36}	7 {37}
22 Feb. 1999	5 {21}	20 {62}
29 Mar. 1999	14 {42}	8 {30}
08 Jun. 1999	1 {4}	8 {24}
08 Nov. 1999	8 {34}	19 {60}

$$SO_{4REM} = \frac{(SO_{4ENT} - SO_{4EXIT})*(Daily\ rate)*(Mol.\ mass)}{(Area\ of\ wetland)} \quad (4)$$

or g m^{-2} day^{-1} = ([moles SO_4 removed]*60*96)/36

where:
SO_{4REM} is the mass removal of sulfate
SO_{4ENT} is the sulfate concentration entering
SO_{4EXIT} is the sulfate concentration exiting
Daily rate is 60 minutes
Molecular mass of sulfate is 96 g
Area of wetland is 36 m^2

Metal mass removal
Generally, zinc and lead removal was consistently greater following the equilibration phase, while

conclusions cannot be made for iron due to insufficient data available for after the equilibration phase (Table 3). Removal of Zn in system 2 ranged between 0.5-70 mg m^{-2} day^{-1}, while for system 1, usually lower removal rates of 0.3-5.7 mg m^{-2} day^{-1} were calculated. Lead was removed by up to 6.6 mg m^{-2} day^{-}1 in system 2 and 5.8 mg m^{-2} day^{-1} in system 1. For iron, up to 2.5 mg m^{-2} day^{-1} was removed. Metal removal rate, as a function of the chemical loading, showed strong correlation coefficients for Zn ($r^2 = 0.99$) and Pb ($r^2 = 0.97$) (O'Sullivan, 2001).

Table 3. Metal removal rates (mg m^{-2} day^{-1}) from water for Zn, Pb and Fe throughout the monitoring period. Values are calculated from differences in concentrations between water entering and exiting the treatment systems (i.e. from the source to the outflow) as a function of the average flow rate (1.5 L min^{-1}), n=5. The calculation is summarized below using Zn as an example (Eqn. 5). Equivalent percentage removals are indicated in parenthesis. Negative values indicate that the metal concentration was higher in water exiting, compared to water entering, on the same sampling date.

SAMPLING	SYSTEM 1	SYSTEM 2
ZINC	*mg m^{-2} day^{-1}*	*mg m^{-2} day^{-1}*
Oct. 1997	0 {0}	1.82 {47}
Nov. 1997	4.9 {95}	0.52 {67}
Jan. 1998	0.3 {50}	0.52 {67}
Nov. 1998	-0.3 {0}	61 {98}
Jan. 1999	1.3 {99}	34 {94}
Jun. 1999	1.3 {71}	8.9 {98}
Mar. 2000	5.7 {67}	70 {99}
LEAD	*mg m^{-2} day^{-1}*	*mg m^{-2} day^{-1}*
Oct. 1997	-0.8 {0}	-0.8 {0}
Nov. 1997	0.8 {33}	0 {0}
Jan. 1998	-0.8 {0}	-0.8 {0}
Nov. 1998	-3.5 {0}	0.8 {25}
Jan. 1999	1.6 {48}	2.5 {60}
Jun. 1999	5.8 {64}	5.8 {47}
Mar. 2000	1.1 {26}	6.6 {50}
IRON	*mg m^{-2} day^{-1}*	*mg m^{-2} day^{-1}*
Nov. 1997	2.5 {85}	2.0 {82}
Jan. 1998	0.2 {99}	0.2 {98}
Nov. 1998	0.4 {29}	2.5 {52}
Jan. 1999	0.4 {29}	2.1 {99}

$$M^{2+}_{REM} = \frac{(M^{2+}_{ENT} - M^{2+}_{EXIT})*(\text{Daily rate})*(\text{Mol. mass})}{(\text{Area of wetland})} \qquad (5)$$

or g m^{-2} day^{-1} = ([moles M^{2+} removed]*60*65)/36

where:
M^{2+}_{REM} is the mass removal of the metal (i.e. Zn)
M^{2+}_{ENT} is the metal concentration entering
M^{2+}_{EXIT} is the metal concentration exiting
Daily rate is 60 minutes
Molecular mass of zinc is 65 g
Area of wetland is 36 m^2

Ecological success and productivity
The treatment systems also demonstrated ecological success through provision of food webs and nesting niches for macroinvertebrates, birds and microbes (Table 4). Both systems were voluntarily colonized by macroinvertebrates, angiosperms, algae, microorganisms and a variety of birds (including *Gallinula chlorpus*). Organic matter contribution to the wetlands during the monitoring period (June 1997 - June 2000) was substantial, particularly by *Typha latifolia* in system 2, which averaged at 700 g m^{-2} yr^{-1} of biomass (data not shown). This vegetation afforded refuge to other visiting wildlife.

Table 4. Number of vegetation, bird and macroinvertebrate species as a function of their colonization time since completion of the systems. Inventories of all species are intended for publication elsewhere, while this data is presented to show the degree of ecological diversity attracted. S. R. (Sulfate Reducing) Bacteria were isolated from (yet indigenous within) substrates.

Type	# Species (identified)	Months post - completion
Angiosperms	5	15
Algae	3	18
Moss	1	12
Avians	3	20
Macroinvert.	34 (21 families)	15
Other Fauna	Frogs (from spawn), rabbits, foxes, shrews	11
S. R. Bacteria	4	12

Discussion

Sulfate removal

Sulfate removal rates were calculated according to Hedin *et al*. (1994), while the equivalent percent removals are also given for comparison. Initially, it was not expected that much sulfate would be removed from the water in the inflow cell, so water sampling in storage tanks and inflow pipes was not conducted. However, early data indicated that substantial sulfate was being removed in the inflow cells, and so sampling regimes were modified to include sampling for sulfate in all locations throughout the water cycle. Lower sulfate removal prior to October 1998 may be explained in part by the initial biogeochemical changes occurring in the systems, which is typical of newly created wetlands (Mitsch & Gossleink, 2000). These changes would have ultimately affected the redox status and therefore the chemistry of the wetland substrates, particularly sulfate dynamics (Engler & Patrick, 1993, Lefroy *et al*., 1993). After the systems had equilibrated (Oct. 1998), sulfate was consistently, although not completely, removed from the water. Post-equilibration phase, sulfate was removed less in the spring (22nd Feb. for system 1 and 29th March for system 2) and summer (8th June) of 1999 than on other dates. This may indicate that sulfate was removed less in the plant growing seasons, compared to non-growing seasons. It is possible that the proliferation of new plant shoots during growing seasons had a negative impact on SRB responsible for biological sulfate reduction, as substrates were locally, albeit temporarily, oxygenated. Oxygen leakage to rhizospheres by wetland plants such as *Typha* and *Phragmites* is well documented, particularly by young shoots (Brix and Schierup, 1990, Wright and Otte, 1999). The activity of sulfate reducing bacteria was not apparently inhibited during winter, possibly due to mild temperatures characteristic of the Irish Atlantic climate. Biologically reduced substrates, coupled with increased organic material renewal, may account for generally more favorable sulfate removal rates seen in non-growing seasons (O'Sullivan*, 2001).

Metal removal

As seen for sulfate, metal removal was calculated from a rate perspective and percent removal was also shown for comparison. Zinc concentrations in water supplied to the treatment systems were higher than background levels but considerably lower than that typical of mine effluent (Gao & Bradshaw, 1995) probably due to substantial precipitation in the tailings storage dam. Additionally, metal concentrations in the water varied considerably depending on the mine works, while water supplied to system 1 was always lower in metal concentrations compared to different water supplied to system 2 (O'Sullivan, 2001). Removal of zinc, lead and iron generally improved after the equilibration phase and removal was greater in system 2 than in system 1. These trends may be attributed to biogeochemical stabilization of the treatment wetlands and to higher concentrations of metals in the water supplied to system 2, respectively. Metal removal, as a function of sampling date, was significant for Zn (system 1; $p<0.05$, system 2; $p<0.001$) and Pb (systems 1 and 2; $p<0.001$), yet this is most likely due to variable chemical loading rather than effects of season. This explanation is supported by the strong correlation between chemical loading and removal seen for Zn and Pb. It is possible that lead concentrations may not have been accurately measured in this research due to insufficient detection capabilities of the instrumentation employed for measuring lead at these low concentrations. Graphite furnace AAS or inductively coupled plasma mass spectrometry is therefore advised.

Contaminant fate

Concentrations of metals significantly accumulated in the upper centimeters of substrates over time (O'Sullivan, 2001) and it is likely that metal sulfides were the dominant form of precipitates (Hammack and Hedin, 1995, Sobolewski, 1996). Metal concentrations in above ground biomass were significantly lower than those associated with below ground biomass (O'Sullivan, 2001), while this is not a recent phenomenon. Predominantly, it was the substrates that operated as a sink for contaminant removal. However, precipitation and related processes are highly dependent on vegetation and thus plant and

substrate processes worked concurrently in removing sulfate and metals from the wastewater.

Ecological function

Wetlands are increasingly recognized for their multi-functional roles, from provision of habitat amenities to landscape restoration, in providing flood control and recreational areas, and more recently, to water quality enhancement via their filtering capacity (Catallo, 1993, Mitsch and Gosselink, 2000). The systems at Tara Mines, although initially designed, were allowed to establish in a 'self-design' process. Roots and rhizomes stabilized the substrate and vegetation afforded refuge to macroinvertebrate and avian communities as well as opportunities for microbial niche establishment. Vegetation also contributed substantial amounts of carbon, renewed annually through plant die-back.

Conclusions

It is apparent that the wetland systems did not reach their treatment capacity (at least for metals) during the course of this monitoring, since an increase in metal levels exiting the systems would have otherwise been expected over time (Mays and Edwards, 2001). The reason for this may be due to insufficient chemical loading (nutrient loading and removal are also correlated in the literature). Additionally, hydrogen sulfide was frequently evolved from the treatment systems and this is indicative of metal exhaustion and/or insufficient loading rates in reduced systems (Hammack and Hedin, 1991). Consequently, long-term treatment of sulfate may be jeopardized since substantial metal cations are required to precipitate high levels of sulfate anions. Despite substantial removal of sulfate from the wastewater within these treatment wetlands, sulfate concentrations exiting were generally not complaint with discharge levels stipulated in the legislation.

It is reported that the performance, such as treatment potential, of ecosystems increases proportionally with ecosystem complexity, hence diversity (Naeem *et al*., 1994). It is therefore reasonable to suggest that the level of diversity provided by the complex array of vegetation, macroinvertebrates and microbes in the constructed wetlands documented here, is high. Furthermore, it may be speculated that successful treatment of the wastewater in these wetland systems was attributed in part to the diversity developed within their ecosystems.

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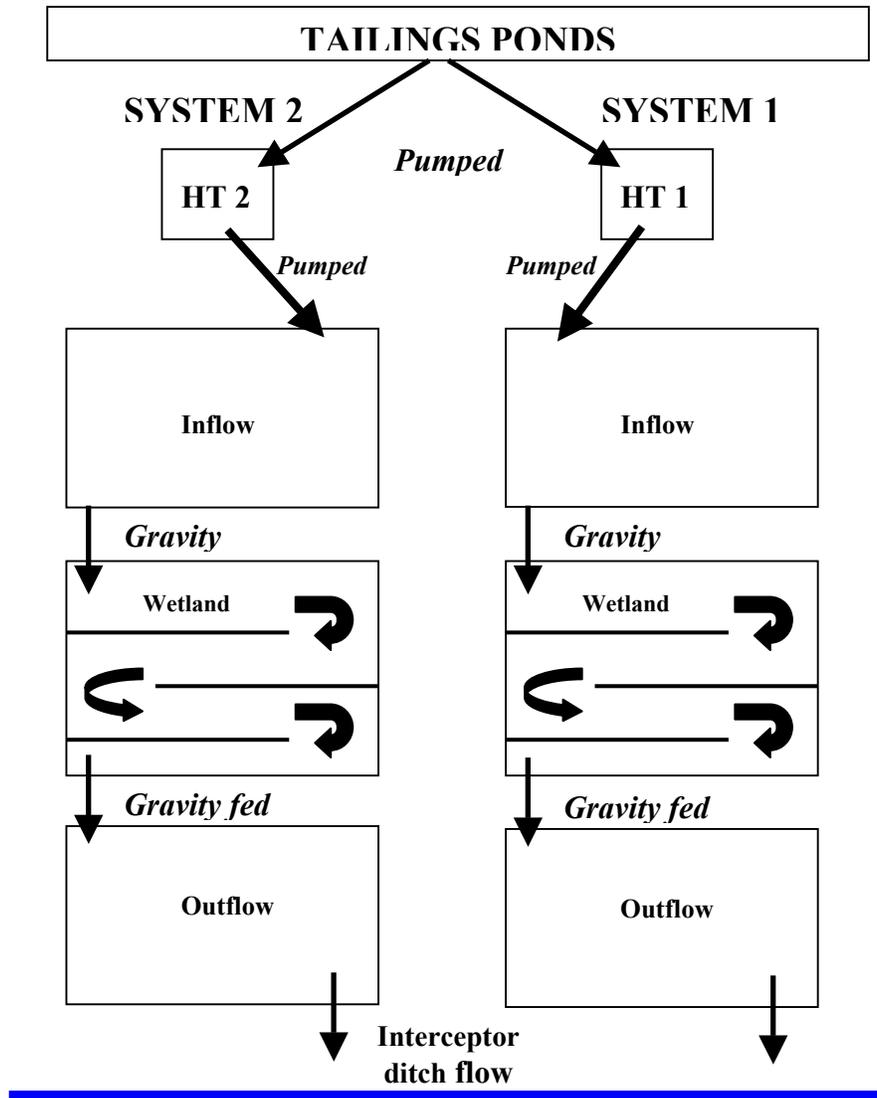
TAILINGS PONDS

SYSTEM 2   SYSTEM 1

Pumped

HT 2   HT 1

Pumped   *Pumped*

Inflow   Inflow

Gravity   *Gravity*

Wetland   Wetland

Gravity fed   *Gravity fed*

Outflow   Outflow

Interceptor ditch flow

Figure 1. Schematic representation of the experimental treatment wetlands at Tara Mines, Ireland. Arrows indicate the flow path of the water, which was pumped from the storage ponds to the header tanks (HT 1 and HT 2) and subsequently to the inflow cells of each system. From there on, water traveling between cells was gravity fed (by head differences between cells) to the wetland cells and then finally to the outflow cells (from John Wiley and Sons Publishers, "Encyclopedia of Water" which will be published in 2004).